

We claim:

1. A multi-layer sheet comprising:

(1) at least one outer layer comprising a propylene polymer material selected from the group consisting of:

(A) *a propylene polymer selected from the group consisting of (i) a propylene homopolymer or a copolymer of propylene and ethylene or (ii) a copolymer of propylene and a 4-8 C alpha-olefin, wherein the polymerized ethylene or polymerized*

alpha-olefin content of the copolymer is not greater than 20%, and,

optionally, about 0.15% to about 0.70% of a nucleating agent,

(b) a graft copolymer comprising a backbone of a propylene polymer material, having graft polymerized thereto polymerized monomers selected from the group consisting of (i) at least one acrylic monomer, (ii) at least one styrenic monomer, and (iii) mixtures of (i) and (ii), and, optionally, about 0.5% to about 1.5% of a nucleating agent, and

(c) an olefin polymer composition comprising:

(i) about 10 parts to about 60 parts by weight of a crystalline propylene homopolymer having an isotactic index greater than 80, or a crystalline copolymer from monomers selected from the group consisting of (1) propylene and ethylene, (2) propylene, ethylene and a C₄-C₈ α-olefin, and (3) propylene and a C₄-C₈ α-olefin, the copolymer having a polymerized propylene content of more than 85% by weight and an isotactic index greater than 85;

(ii) about 5 parts to about 25 parts by weight of a copolymer of ethylene and propylene or a C₄-C₈ α-olefin that is insoluble in xylene at ambient temperature; and

(iii) about 30 parts to about 70 parts by weight of an elastomeric copolymer from monomers selected from the group consisting of (1) ethylene and propylene, (2) ethylene, propylene, and a C₄-C₈ α-

olefin, and (3) ethylene and a C₄-C₈ α-olefin, the copolymer optionally containing about 0.5% to about 10% by weight of a polymerized diene, and containing less than 70% by weight of polymerized ethylene and being soluble in xylene at ambient temperature and having an intrinsic viscosity measured in decahydronaphthalene at 135°C of about 1.5 to about 4.0 dl/g; the total of (ii) and (iii), based on the total olefin polymer composition being from about 50% to about 90%, and the weight ratio of (ii)/(iii) being less than 0.4, wherein the composition is prepared by polymerization in at least two stages and has a flexural modulus of less than 150 MPa;

(2) at least one layer comprising alternating ^{sub-} layers of ^(a) a propylene polymer material selected from the group consisting of ⁽ⁱ⁾ a propylene homopolymer and ⁽ⁱⁱ⁾ a copolymer of propylene and ethylene ^{and (iii) a copolymer of propylene and} of a 4-8 C alpha-olefin, wherein the polymerized ethylene or polymerized alpha-olefin content of the copolymer is not greater than 20%, and the melt flow rate of the propylene polymer material in layer (2) is higher than the melt flow rate of the propylene polymer material in layer (2), ⁽¹⁾ and (b) glass fiber mats having a weight per unit area of about 0.5 to about 4.5 oz/ft², with the propylene polymer material as the top and bottom layers, wherein layer (1) has a thickness of about 0.001 inch to about 0.25 inch.

2. The multi-layer sheet of claim 1 wherein the polymerized monomers in the graft copolymer (b) of layer (1) are a mixture of methyl methacrylate and methyl acrylate.

3. The multi-layer sheet of claim 1 wherein the propylene polymer material that is the backbone polymer of the graft copolymer (b) in layer (1) is selected from the group consisting of:

(a) a homopolymer of propylene having an isotactic index greater than 80;

(b) a copolymer of propylene and an olefin selected from the group consisting of ethylene and 4-10 C alpha-olefins, provided that when the olefin is ethylene, the

maximum polymerized ethylene content is about 10% and when the olefin is a 4-10 C alpha-olefin, the maximum polymerized content thereof is about 20% by weight, the copolymer having an isotactic index greater than 85;

- 5 (c) a terpolymer of propylene and two olefins selected from the group consisting of ethylene and 4-8 C alpha-olefins, provided that the maximum polymerized 4-8 C alpha-olefin content is 20% by weight, and, when ethylene is one of the olefins, the maximum polymerized ethylene content is 5% by weight, the terpolymer having an isotactic index greater than 85;

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(d) an olefin polymer composition comprising:

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(i) about 10% to about 60% by weight of a propylene homopolymer having an isotactic index greater than 80, or a copolymer of monomers selected from the group consisting of (1) propylene and ethylene, (2) propylene, ethylene and a 4-8 C alpha-olefin, and (3) propylene and a 4-8 C alpha-olefin, the copolymer having a polymerized propylene content of more than 85% by weight and an isotactic index greater than 85;

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(ii) about 5% to about 25% by weight of a copolymer of ethylene and propylene or a 4-8 C alpha-olefin that is insoluble in xylene at ambient temperature; and

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(iii) about 30% to about 70% by weight of an elastomeric copolymer of monomers selected from the group consisting of (1) ethylene and propylene, (2) ethylene, propylene, and a 4-8 C alpha-olefin, and (3) ethylene and a 4-8 C alpha-olefin, the copolymer optionally containing about 0.5% to about 10% by weight of a polymerized diene and containing less than 70% by weight of polymerized ethylene and being soluble in xylene at ambient temperature and having an intrinsic viscosity measured in decahydronaphthalene at 135°C of about 1.5 to about 4.0 dl/g,

wherein the total amount of (ii) and (iii), based on the total olefin polymer composition, is about 50% to about 90%, the weight ratio of (ii)/(iii) is less than 0.4, and the composition is prepared by polymerization in at least two stages and has a flexural modulus of less than 150 MPa; and

5 (e) a thermoplastic olefin comprising:

(i) about 10% to about 60% of a propylene homopolymer having an isotactic index greater than 80, or a copolymer of monomers selected from the group consisting of (1) ethylene and propylene, (2) ethylene, propylene and a 4-8 C alpha-olefin, and (3) ethylene and a 4-8 C alpha-olefin, the
10 copolymer having a polymerized propylene content greater than 85% and an isotactic index of greater than 85;

(ii) about 20% to about 60% of an amorphous copolymer from monomers selected from the group consisting of (1) ethylene and propylene, (2) ethylene, propylene, and a 4-8 C alpha-olefin, and (3) ethylene and a 4-8 C
15 alpha-olefin, the copolymer optionally containing about 0.5% to about 10% of a polymerized diene, and containing less than 70% polymerized ethylene and being soluble in xylene at ambient temperature; and

(iii) about 3% to about 40% of a copolymer of ethylene and propylene or a 4-8 C alpha-olefin that is insoluble in xylene at ambient temperature,
20 wherein the thermoplastic olefin has a flexural modulus of greater than 150 but less than 1200 MPa.

4. The multi-layer sheet of claim 3 wherein the propylene polymer material is a propylene homopolymer.

5. A composite material comprising a central core of synthetic resin material
25 sandwiched between two of the multi-layer sheets of claim 1, wherein layer (1) is always on the outside of the composite material.

6. The composite material of claim 5 wherein the core is a foam made from a polyolefin having strain hardening elongational viscosity.

7. The composite material of claim 5 wherein the core is a polyolefin honeycomb material.

8. A process for making a multi-layer sheet comprising:

(1) extruding a sheet comprising a propylene polymer material selected

from the group consisting of:

a propylene polymer selected from the group consisting of (i)

(a) *(i)* a propylene homopolymer *or (ii)* a copolymer of propylene and

ethylene *or a 4-8 C alpha-olefin, wherein the polymerized ethylene*

and (ii) a copolymer of propylene and or polymerized alpha-olefin content of the copolymer is not greater

than 20%, and, optionally about 0.15% to about 0.70% of a

nucleating agent,

(b) a graft copolymer comprising a backbone of a propylene

polymer material, having graft polymerized thereto polymerized

monomers selected from the group consisting of (i) at least one

acrylic monomer, (ii) at least one styrenic monomer, and (iii)

mixtures of (i) and (ii), and, optionally, about 0.5% to about 1.5%

of a nucleating agent, and

(c) an olefin polymer composition comprising:

(i) about 10 parts to about 60 parts by weight of a

crystalline propylene homopolymer having an isotactic index

greater than 80, or a crystalline copolymer from monomers

selected from the group consisting of (1) propylene and

ethylene, (2) propylene, ethylene and a C₄-C₈ α-olefin, and

(3) propylene and a C₄-C₈ α-olefin, the copolymer having a

polymerized propylene content of more than 85% by weight

and an isotactic index greater than 85;

(ii) about 5 parts to about 25 parts by weight of a

copolymer of ethylene and propylene or a C₄-C₈ α-olefin

that is insoluble in xylene at ambient temperature; and

(iii) about 30 parts to about 70 parts by weight of an elastomeric copolymer from monomers selected from the group consisting of (1) ethylene and propylene, (2) ethylene, propylene, and a C₄-C₈ α-olefin, and (3) ethylene and a C₄-C₈ α-olefin, the copolymer optionally containing about 0.5% to about 10% by weight of a polymerized diene, and containing less than 70% by weight of polymerized ethylene and being soluble in xylene at ambient temperature and having an intrinsic viscosity measured in decahydro-naphthalene at 135°C of about 1.5 to about 4.0 dl/g; the total of (ii) and (iii), based on the total olefin polymer composition being from about 50% to about 90%, and the weight ratio of (ii)/(iii) being less than 0.4, wherein the composition is prepared by polymerization in at least two stages and has a flexural modulus of less than 150 MPa;

the sheet of propylene polymer material having a thickness of about 0.001 inch to about 0.25 inch,

(2) forming a sheet by laying-up alternate layers of (a) a sheet of a propylene polymer material selected from the group consisting of (i) a propylene homopolymer and (ii) a copolymer of propylene and ethylene or a 4-8 C alpha-olefin, wherein the polymerized ethylene or polymerized alpha-olefin content of the copolymer is not greater than 20%, the melt flow rate of the propylene polymer material in layer (2) is higher than the melt flow rate of the propylene polymer material in layer (1), and the sheet has a thickness of about 0.01 to about 0.1 inch, and (b) glass fiber mats having a weight per unit area of about 0.5 to about 4.5 oz/ft², that have been preheated to a temperature of about 140° to about 200°C, with the sheet of propylene polymer material as the top and bottom layers,

compressing the layers at a pressure of at least 300 psi, and then cooling while maintaining this pressure, and

(3) joining the sheets produced in steps (1) and (2) by applying heat to both sheets at a temperature sufficient to soften the surfaces to be joined, while simultaneously applying pressure to the sheet produced in step (1).

9. The process of claim 8 wherein the polymerized monomers in the graft copolymer (b) of layer (1) are a mixture of methyl methacrylate and methyl acrylate.

10. The process of claim 8 wherein the propylene polymer material that is the backbone polymer of the graft copolymer (b) in layer (1) is selected from the group consisting of:

(a) a homopolymer of propylene having an isotactic index greater than 80;

(b) a copolymer of propylene and an olefin selected from the group consisting of ethylene and 4-10 C alpha-olefins, provided that when the olefin is ethylene, the maximum polymerized ethylene content is about 10% and when the olefin is a 4-10 C alpha-olefin, the maximum polymerized content thereof is about 20% by weight, the copolymer having an isotactic index greater than 85;

(c) a terpolymer of propylene and two olefins selected from the group consisting of ethylene and 4-8 C alpha-olefins, provided that the maximum polymerized 4-8 C alpha-olefin content is 20% by weight, and, when ethylene is one of the olefins, the maximum polymerized ethylene content is 5% by weight, the terpolymer having an isotactic index greater than 85;

(d) an olefin polymer composition comprising:

(i) about 10% to about 60% by weight of a propylene homopolymer having an isotactic index greater than 80, or a copolymer of monomers selected from the group consisting of (1) propylene and ethylene, (2) propylene,

ethylene and a 4-8 C alpha-olefin, and (3) propylene and a 4-8 C alpha-olefin, the copolymer having a polymerized propylene content of more than 85% by weight and an isotactic index greater than 85;

(ii) about 5% to about 25% by weight of a copolymer of ethylene and propylene or a 4-8 C alpha-olefin that is insoluble in xylene at ambient temperature; and

(iii) about 30% to about 70% by weight of an elastomeric copolymer of monomers selected from the group consisting of (1) ethylene and propylene, (2) ethylene, propylene, and a 4-8 C alpha-olefin, and (3)

ethylene and a 4-8 C alpha-olefin, the copolymer optionally containing about 0.5% to about 10% by weight of a polymerized diene and containing less than 70% by weight of polymerized ethylene and being soluble in xylene at ambient temperature and having an intrinsic viscosity measured in decahydronaphthalene at 135°C of about 1.5 to about 4.0 dl/g,

wherein the total amount of (ii) and (iii), based on the total olefin polymer composition, is about 50% to about 90%, the weight ratio of (ii)/(iii) is less than 0.4, and the composition is prepared by polymerization in at least two stages and has a flexural modulus of less than 150 MPa; and

(e) a thermoplastic olefin comprising:

(i) about 10% to about 60% of a propylene homopolymer having an isotactic index greater than 80, or a copolymer of monomers selected from the group consisting of (1) ethylene and propylene, (2) ethylene, propylene and a 4-8 C alpha-olefin, and (3) ethylene and a 4-8 C alpha-olefin, the copolymer having a polymerized propylene content greater than 85% and an isotactic index of greater than 85;

(ii) about 20% to about 60% of an amorphous copolymer from monomers selected from the group consisting of (1) ethylene and propylene, (2) ethylene, propylene, and a 4-8 C alpha-olefin, and (3) ethylene and a 4-8 C

alpha-olefin, the copolymer optionally containing about 0.5% to about 10% of a polymerized diene, and containing less than 70% polymerized ethylene and being soluble in xylene at ambient temperature; and

(iii) about 3% to about 40% of a copolymer of ethylene and propylene or a 4-8 C alpha-olefin that is insoluble in xylene at ambient temperature,

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wherein the thermoplastic olefin has a flexural modulus of greater than 150 but less than 1200 MPa.

11. The process of claim 10 wherein the propylene polymer material is a propylene homopolymer.